



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of: **Tetsuro YAMATE**

Group Art Unit: **1712**

Application Number: **10/820,844**

Examiner: **Daniel S. Metzmaier**

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For: **CHEMILUMINESCENT COMPOSITION**

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DECLARATION UNDER 37 CFR §1.132

Commissioner for Patents
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June 2, 2008

Sir:

I, Tetsuro Yamate, hereby declare and state that:

(1) I am an inventor of the invention described in US Serial Number 10/820,844. I have been engaged in research of a chemiluminescent composition and chemiluminescent device at Lumica Corporation from 1993 to the present.

(2) I am familiar with the contents of the United States Patent Application Serial No. 10/820,844 filed on April 9, 2004. I have read Cranor (US 7,052,631 or 2003/0102467) and Omniglow (WO94/19421).

(3) I prepared a declaration under 37 CFR §1.132 ("1st Declaration") which I prepared on June 8, 2007 and submitted on June 12, 2007.

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(4) I prepared a declaration under 37 CFR §1.132 ("2nd Declaration") which I prepared on September 29, 2007 and submitted on September 29, 2007.

(5) I have read the outstanding Office Action mailed on January 2, 2008. In order to show unexpected results of the present invention over Cranor and Omniglow, I conducted Test 3-8 in this Declaration. The numbers of Samples and Tables in this Declaration continues from the 2nd Declaration.

(6) Test 3

Using butyl benzoate (BtB) and acetyltributyl citrate (ATBC), chemiluminescent compositions including 1-chloro bis(phenylethynyl)anthracene (1-c BPEA) and bis (2,4,5-trichloro carbopentoxyphehyl) oxalate (CPPO) were prepared in the same manner as described in the specification to obtain Samples 6-7 as follow:

0.0074 mol of 1-chloro bis(phenylethynyl)anthracene (1-c BPEA) and 0.08 mol of bis (2,4,5-trichloro carbopentoxyphehyl) oxalate (CPPO) were added to 1 L of butyl benzoate (BtB), which was then heated to obtain Sample 6.

0.0074 mol of 1-chloro bis(phenylethynyl)anthracene (1-c BPEA) and 0.08 mol of bis (2,4,5-trichloro carbopentoxyphehyl) oxalate (CPPO) were added to 1 L of acetyltributyl citrate (ATBC), which were then heated to obtain Sample 7.

An oxidizing liquid including hydrogen peroxide was prepared in the same manner as the

description at page 4, lines 14-16 of the specification of the present application.

0.42 cc of the oxidizing liquid was mixed with 0.84 cc of Samples 6-7 to induce luminescence.

The results of the luminescent intensity are shown in Table 4 below.

Table 4

time (minutes)	2	15	60	120	180	240	300	360
Sample 6 (BtB 100%)	22480	20600	14110	5673	3071	1585	682	462
	62%	84%	94%	82%	94%	93%	74%	89%
Sample 7 (ATBC 100%)	36060	24380	14980	6888	3281	1712	919	520
	100%	100%	100%	100%	100%	100%	100%	100%

Measurement at 23°C

Luminescent Intensity: candela (mcd/m²) (measured by a luminance meter available from Minolta Camera Co. Ltd., Japan)

The results listed in Table 4 shows that the luminescent intensity of Sample 6 was lower than Sample 7 by 6% to 38%. In particular, the luminescent intensity of Sample 6 was lower than Sample 7 by 38% at the time of 2 minutes. The results were unexpected. ATBC is advantageous in using a solvent for the chemiluminescent composition compared with butyl benzoate.

(7) Test 4

Samples 6-7 were kept in a glass vessel at a temperature of 60°C for a period of one week. Then, the concentration of CPPO in the Samples was measured by using a high rate liquid chromatograph, Hitachi L7400.

As results, the concentration of CPPO of Sample 6 was decreased by 17%, whereas the concentration of CPPO of Sample 7 was decreased by 1%.

The decomposition of CPPO of Sample 6 was much more than that of Sample 7. The results are unexpected. Acetyltributyl citrate (ATBC) is advantageous in the storage properties compared with butyl benzoate.

(8) Test 5

Samples 6-7 were stored in a glass vessel at a temperature of 60°C for a period of one week. Then, 0.42 cc of the oxidizing liquid was mixed with 0.84 cc of the stored Samples 6-7 to induce luminescence. The results of the luminescent intensity are listed in Table 5 below.

Table 5

time (minutes)	2	15	60	120	180	240	300	360
Sample 6 using BtB	23100	20890	14160	4906	1641	487	163	91
(Compared with Sample 7)	65%%	87%	97%	75%	51%	29%	19%	18%
Sample 7 using ATBC	35420	24100	14620	6523	3195	1693	878	508
	100%	100%	100%	100%	100%	100%	100%	100%

Measurement at 23°C

Luminescent Intensity: candela (mcd/m²) (measured by a luminance meter available from Minolta Camera Co. Ltd., Japan).

The results listed in Table 5 shows that the luminescence of Sample 6 was lower than Sample 7. In particular, the luminescence of Sample 6 was significantly lower than Sample 7 at the time of 2minuts, and at the time after 180 minutes. The results were unexpected. ATBC is advantageous in the storage properties compared with butyl benzoate (BtB).

(9) Test 6

The following solutions were provided:

Propylene glycol dibenzoate (PGDB) /Acetyltributyl citrate (ATBC) (50/50);

ATBC/Benzyl Benzoate (BeB) (100%/0%);

ATBC/Benzyl Benzoate (BeB) (90%/10%);

ATBC/Benzyl Benzoate (BeB) (80%/20%);

ATBC/Benzyl Benzoate (BeB) (70%/30%); and

ATBC/Benzyl Benzoate (BeB) (60%/40%).

0.0074 mol of 1-chloro bis(phenylethynyl)anthracene (1-c BPEA) and 0.08 mol of bis (2,4,5-trichloro carbopentoxyphehyl) oxalate (CPPO) were added to 1 L of Propylene glycol dibenzoate (PGDB) /Acetyltributyl citrate (ATBC) (50/50), which was then heated to obtain Sample 8.

0.0074 mol of 1-chloro bis(phenylethynyl)anthracene (1-c BPEA) and 0.08 mol of bis (2,4,5-trichloro carbopentoxyphehyl) oxalate (CPPO) were added to 1 L of ATBC (100%), which was then heated to obtain Sample 9.

0.0074 mol of 1-chloro bis(phenylethynyl)anthracene (1-c BPEA) and 0.08 mol of bis (2,4,5-trichloro carbopentoxyphehyl) oxalate (CPPO) were added to 1 L of ATBC/Benzyl Benzoate (BeB) (90%/10%), which was then heated to obtain Sample 10.

0.0074 mol of 1-chloro bis(phenylethynyl)anthracene (1-c BPEA) and 0.08 mol of bis

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(2,4,5-trichloro carbopentoxyphenyl) oxalate (CPPO) were added to 1 L of ATBC/Benzyl Benzoate (BeB) (80%/20%), which was then heated to obtain Sample 11.

0.0074 mol of 1-chloro bis(phenylethynyl)anthracene (1-c BPEA) and 0.08 mol of bis (2,4,5-trichloro carbopentoxyphenyl) oxalate (CPPO) were added to 1 L of ATBC/Benzyl Benzoate (BeB) (70%/30%), which was then heated to obtain Sample 12.

0.0074 mol of 1-chloro bis(phenylethynyl)anthracene (1-c BPEA) and 0.08 mol of bis (2,4,5-trichloro carbopentoxyphenyl) oxalate (CPPO) were added to 1 L of ATBC/Benzyl Benzoate (BeB) (60%/40%), which was then heated to obtain Sample 13.

0.0074 mol of 1-chloro bis(phenylethynyl)anthracene (1-c BPEA) and 0.08 mol of bis (2,4,5-trichloro carbopentoxyphenyl) oxalate (CPPO) were added to 1 L of ATBC/Benzyl Benzoate (BeB) (50%/50%), which was then heated to obtain Sample 14.

Then, 0.42 cc of the oxidizing liquid was mixed with 0.84 cc of the Samples 8-14 to induce luminescence. The results of the decay of the luminescent intensity are shown in Table 6.

Table 6

time(minutes)	2	15	60	120	180	240	300	360
Sample 8	21310	15930	9970	5406	3803	1981	1113	657
PGDB/ATBC (50/50)	59%	65%	66%	78%	116%	116%	121%	126%
Sample 9	36060	24380	14980	6888	3281	1712	919	520
ATBC/BeB (100%/0%)	100%	100%	100%	100%	100%	100%	100%	100%
Sample 10	35990	22990	14790	6900	3213	1790	903	512
ATBC/BeB (90%/10%)	100%	94%	99%	100%	98%	105%	98%	98%
Sample 11	34000	22190	14540	6830	3183	1707	890	503
ATBC/BeB (80%/20%)	94%	91%	97%	99%	97%	100%	97%	97%
Sample 12	33430	21920	14000	6645	3129	1678	920	497
ATBC/BeB (70%/30%)	93%	90%	93%	96%	95%	98%	100%	96%
Sample 13	30780	19870	13280	6239	3138	1622	884	492
ATBC/BeB (60%/40%)	85%	82%	87%	91%	96%	95%	96%	95%
Sample 14	29970	19130	12870	5829	3033	1620	877	494
ATBC/BeB (50%/50%)	83%	78%	86%	85%	92%	95%	95%	95%

Measurement at 23°C

Luminescent Intensity: candela (mcd/m²) (measured by a luminance meter available from Minolta Camera Co. Ltd., Japan)

(i) Comparing Sample 8 with Sample 14, Sample 14 improved the luminescent intensity especially at the time of 2 to 120 minutes. The results obtained by replacement of glycol dibenzoate (PGDB) with benzyl benzoate (BeB) were unexpected.

(ii) Comparing Sample 8 with Samples 9-12, the luminescent intensity of Samples 9-12 was much more improved than that of Sample 8 especially at the time of 2 to 120 minutes. The results obtained by adding benzyl benzoate at an amount of 30% by volume or less were unexpected.

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(10) Test 7

Samples 8-14 were kept in a glass vessel at a temperature of 60°C for a period of one week. Then, the concentration of CPPO in the Samples was measured using a high rate liquid chromatograph, Hitachi L7400.

The results were as follows:

The concentration of CPPO in Sample 8 was decreased by 10%. The concentrations of CPPO in Samples 9-14 were decreased by 1%, 1%, 1%, 3%, 4% and 5%, respectively.

The decomposition of CPPO in Sample 8 was more than that in Samples 9-14. The decomposition of CPPO in Samples 9-12 were unexpectedly inhibited compared with that in Sample 8.

(11) Test 8

Samples 8-14 were stored at a temperature of 60°C for a period of one week. Then, 0.42 cc of the oxidizing liquid was mixed with 0.84 cc of the stored Samples 8-14 to induce luminescence. The results of the decay of the luminescent intensity were shown in Table 7.

Table 7

time(minutes)	2	15	60	120	180	240	300	360
Sample 8	18880	15270	10240	5274	3272	1491	747	383
PGDB/ATBC (50/50)	53%	63%	70%	81%	102%	88%	85%	75%
Sample 9	35420	24100	14620	6523	3195	1693	878	508
ATBC (100%)	100%	100%	100%	100%	100%	100%	100%	100%
Sample 10	32640	21930	13940	6422	2903	1693	842	485
ATBC/BeB (90%/10%)	92%	91%	95%	98%	91%	100%	96%	95%
Sample 11	31320	20690	13540	6270	2754	1493	812	455
ATBC/BeB (80%/20%)	88%	86%	93%	96%	86%	88%	92%	90%
Sample 12	29300	21230	13680	6396	2764	1371	744	382
ATBC/BeB (70%/30%)	83%	88%	94%	98%	87%	81%	85%	75%
Sample 13	23860	18430	13160	6129	2684	1236	602	213
ATBC/BeB (60%/40%)	67%	76%	90%	94%	84%	73%	69%	42%
Sample 14	24460	18560	12990	5734	2667	1199	487	183
ATBC/BeB (50%/50%)	69%	77%	89%	88%	83%	71%	55%	36%

Measurement at 23°C

Luminescent Intensity: candela (mcd/m²) (measured by a luminance meter available from Minolta Camera Co. Ltd., Japan).

(i) Comparing Sample 8 with Sample 14, Sample 14 improved the luminescent intensity especially at the time of 2 to 120 minutes. The results obtained by replacement of glycol dibenzoate (PGDB) with benzyl benzoate (BeB) were unexpected. Benzyl benzoate was advantageous in the storage properties compared with glycol dibenzoate (PGDB).

(ii) Comparing Sample 8 with Samples 9-12, the luminescent intensity of Samples 9-12 was improved compared with that of Sample 8 especially at the time of 2 to 120 minutes. The results obtained

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by adding BeB at an amount of 30% by volume or less were unexpected. Benzyl benzoate including at an amount of 30% by volume or less was advantageous in the storage properties compared with glycol dibenzoate (PGDB).

(12) I state regarding the results in the 1st Declaration as follows: Comparison of Sample B (acetyltributyl citrate) with Samples A and C shows an unexpected result that acetyltributyl citrate is used as a solvent for an oxalate/fluorescent composition to exhibit a performance compatible with dimethyl phthalate.

(13) I state regarding the results in the 2nd Declaration as follows: The fluorescent composition including acetyltributyl citrate (Sample 2) was superior to that including acetyl triethyl citrate or triethyl citrate. The results of the present invention presented in the 2nd Declaration are unexpected.

(14) I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like are punishable by fine or imprisonment, or both, under the laws of the United State and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

<u>Tetsuro Yamate</u>	<u>June 2, 2008</u>
Tetsuro Yamate	Date